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# A climatology of $^{7}$Be in surface air in European Union

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ABSTRACT

This study presents a European-wide analysis of the spatial and temporal distribution of the cosmogenic isotope $^{7}$Be in surface air. This is the first time that a long term database of 34 sampling sites that regularly provide data to the Radioactivity Environmental Monitoring (REM) network, managed by the Joint Research Centre (JRC) in Ispra, is used. While temporal coverage varies between stations, some of them have delivered data more or less continuously from 1984 to 2011. The station locations were considerably heterogeneous, both in terms of latitude and altitude, a range which should ensure a high degree of representativeness of the results.

The mean values of $^{7}$Be activity concentration presented a spatial distribution value ranging from 2.0 to 5.4 mBq/m$^{3}$ over the European Union. The results of the ANOVA analysis of all $^{7}$Be data available indicated that its temporal and spatial distributions were mainly explained by the location and characteristic of the sampling sites rather than its temporal distribution (yearly, seasonal and monthly). Higher $^{7}$Be concentrations were registered at the middle, compared to high-latitude, regions. However, there was no correlation with altitude, since all stations are sited within the atmospheric boundary layer. In addition, the total and yearly analyses of the data indicated a dynamic range of $^{7}$Be activity for each solar cycle and phase (maximum or minimum), different impact on stations having been observed according to their location. Finally, the results indicated a significant seasonal and monthly variation for $^{7}$Be activity concentration across the European Union, with maximum concentrations occurring in the summer and minimum in the winter, although with differences in the values reached.

The knowledge of the horizontal and vertical distribution of this natural radionuclide in the atmosphere is a key parameter for modelling studies of atmospheric processes, which are important phenomena to be taken into account in the case of a nuclear accident.



## 1. Introduction

The cosmogenic isotope Berillium-7 ($^{7}$Be) is produced by spallation reactions through interaction of cosmic rays with atmospheric molecules of nitrogen, oxygen and carbon (Masarik and Beer, 1999). In detail, the 75% production occurs in the stratosphere while the 25% is produced in the troposphere, and particularly in the upper troposphere (Johnson and Viezee, 1981; Usoskin and Kovaltsov, 2008). Its production is primarily controlled by latitude and altitude but also varies as a function of solar particle and magnetic flux and the geomagnetic field strength (Lal and Peters, 1967; Usoskin et al., 2009a, b).

Once produced, $^{7}$Be is rapidly adsorbed onto aerosol particles in the stratosphere and troposphere and further transported to the Earth's surface by atmospheric vertical mixing. In this sense, $^{7}$Be undergoes rapid association onto submicron-sized aerosol particles in the accumulation mode ($N_{ACC}$) (particles with diameters within (0.1–1) μm size range) (Gaffney et al., 2004; Ioannidou et al., 2005), which is the reason why $^{7}$Be is removed by (wet, and secondarily dry) deposition. Several factors affect the distribution of $^{7}$Be in surface air, such as the air mass exchange between stratosphere and troposphere, vertical transport in the troposphere, horizontal transport from subtropics and mid-latitudes to tropics and Polar Regions. All these parameters introduce complexity in the evaluation of the $^{7}$Be activity in surface air.

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Many studies have described $^{7}$Be concentrations in surface air in local sampling stations in Europe: Azahra et al., 2003, 2004; Lozano et al., 2011, Dueñas et al., 2011 and Piñero Garcia et al., 2012 in Spain, Likuku, 2006 and Daish et al., 2005 in England; Leppänen et al., 2010, 2012; Leppänen and Paatero, 2013 in Finland; Cannizzaro et al., 2004; Tositti et al., 2014 in Italy; Papastefanou and Ioannidou 1991 and Ioannidou et al., 2005 in Greece; Carvalho et al., 2013 in Portugal; Pham et al., 2011 in Monaco, Steinmann et al., 2013 in Switzerland; Todorovic et al., 1999, 2005 in Yugoslavia. In the same line, several studies have comprised the analysis of regional distribution of $^{7}$Be in Europe, e.g. Leppänen and Paatero, 2013 analysed the surface air $^{7}$Be concentrations in Finland according to solar cycle, Kulan et al., 2006 performed the European analysis of $^{7}$Be taking five monitoring stations as reference, and Gerasopoulos et al., 2001 and Tositti et al., 2004 carried out a comparisons of $^{7}$Be measurements at four high-altitude stations in Europe.

However, a comprehensive analysis of $^{7}$Be distribution at European level has not yet been undertaken. This kind of analysis of $^{7}$Be is crucial to better understand its global distribution. Improving the understanding of the $^{7}$Be temporal and spatial atmospheric distribution is a powerful means to study changes in atmospheric processes (such as dry and wet deposition), which are important processes to be taken into account in the case of a nuclear accident. In addition, better understanding these phenomena may also make it easier to validate global circulation models. In this sense, Brost et al., 1991, Koch and Mann, 1996 and Koch et al., 1996, Land and Feichter 2003, and Liu et al., 2001, 2004 report simulations and comparisons with $^{7}$Be activity concentrations.

Considering this gap, the current study analyses the behaviour of $^{7}$Be in surface air in the European Union (EU) (from 35° to 72°N and 20°W–40°E), using data collected by the Radioactivity Environmental Monitoring (REM) group in order to characterize well the temporal and spatial variations of each of the locations that provides data to REM on a regular basis. To increase the representativeness and validity of the results, we cover a relatively long time period (from 1984 onwards, given data availability, essentially starting from the year in which a country became a full Member State of the EU), with a tightly spaced sampling interval (daily to monthly), using a large set of monitoring locations from the sparse monitoring network around Europe (34 sampling stations in total). Briefly, this network is integrated by a number of representative locations in each EU country with high-sensitivity measurements.

Following a brief description of the REM Database and $^{7}$Be measurement procedures, the following sections will show results from ANOVA analysis to define the variation of $^{7}$Be concentrations explained by spatial (station) and temporal (yearly, seasonal and monthly) basis. In the rest of the sections, statistical results obtained from the overall period will be presented. The data analysis of the variability of $^{7}$Be data as averaged values in a yearly, seasonal and monthly basis is performed to fully account for the spatial and temporal variability in surface air $^{7}$Be concentrations. In addition, the impact of the 11-y solar modulation on the $^{7}$Be concentrations in air is addressed in the present study.

In this work, we aim to address the following research issues:

- To analyse the distribution of surface air $^{7}$Be concentrations in Europe;
- To investigate the temporal variability (yearly, seasonal and monthly);
- To investigate the impact of solar cycles on surface air $^{7}$Be concentrations;

## 2. The REM database

The quantity and diversity of environmental radioactivity data produced after the Chernobyl accident demonstrated the need for an effective system to integrate, store and retrieve them. The online Radioactivity Environmental Monitoring (REM) Database, supported by the Radioactivity Environmental Monitoring group of the Institute for Transuranium Elements (ITU) of the Joint Research Centre (JRC), was established in 1988 with the aims to: 1) facilitate the exchange of information between the EU Member States and the European Commission (EC); and 2) to collect and store environmental radioactivity data produced in the aftermath of the Chernobyl accident for scientific study and for obtaining a European overview of the contamination situation.

The REM Database was conceived as a series of data records, each one containing a single measurement of a single radionuclide on a single sample. Under the terms of Article 36 of the Euratom Treaty, Member States shall periodically communicate to the Commission information on environmental radioactivity levels. Nowadays, REM contains a unique collection of environmental radioactivity measurements from a wide number of different sources, media and countries from 1984 onwards (with some date being even older). Currently, the REM Database contains more than 2 million measurements of both environmental samples and foodstuffs, spanning sample types such as air, deposition, water, milk, meat and vegetables are the best represented.

Therefore, REM is a unique resource of consistently structured information of clear benefit to anyone wishing to handle, analyse and compare environmental radioactivity data across organisational and country boundaries. Applications may vary from simple mapping, statistical interpretation and trend analysis, to model validation of behaviour and transport systems and dose assessments. More information on the REM Database can be found on its website: http://rem.jrc.ec.europa.eu/.

## 3. Sampling and analytical methods

The measurements of $^{7}$Be were made on air filters collected at sampling networks indicated in Table 1. The local geographical location of each sampling device ensures that the sample obtained is representative of the air around. Airborne particulate sampling is carried out by pumping air through filters at a flow rate of several hundred cubic meters per day. Further information on the procedure to collect aerosol samples from ground level air at each network, as well as the list of National Competent Authorities can be found in the Appendix. Individual radionuclide analyses are performed daily, weekly, monthly or quarterly.

Table 1 presents the temporal coverage of data in each station. We have selected data records having end in December 2011 and with at least five years duration. The begin time for those record is essentially conditioned by the year in which that country became a Member States, assuming regular data delivery since then. In some cases, we have records spanning no less than 25 years. The sampling availability ranges from 4297 to 10 observations. This large variability in the database of each station is due to differently spaced sampling intervals (daily to yearly). Considering this large difference in the temporal resolution of the data, and the temporal gaps identified in some of the sampling sites, a quality criterion was applied to ensure the representativeness of the results. Our criterion requires that each station have a completed year of $^{7}$Be measurements (with a minimum of 12 monthly measurements) with the aim to cover the monthly variability at least during a year in each sampling site. Considering the whole sampling period in each station, each monthly value was calculated by taking monthly, weekly or daily measurements registered in the corresponding

**Table 1**
Station name, coordinates (Latitude, longitude and height above sea level), sampling period and the number of operability sampling periods together with the average of air surface $^{7}$Be activity concentration at all the stations of the REM network.

| Stations | LAT | LON | Altitude (m a.s.l) | Sampling period | Operability (samplings) | Mean + SD (mBq/m3) |
|---|---|---|---|---|---|---|
| Ivalo | 68.64 | 27.57 | 130 | Feb 1987–Dec 2011 | 1189 | 2.0 ± 0.9 |
| Umea | 63.85 | 20.34 | 45 | Jan 1995–Dec 2011 | 875 | 2.0 ± 0.9 |
| Helsinki | 60.21 | 25.06 | 12 | Jan 1987–Dec 2011 | 4297 | 2.2 ± 1.3 |
| Kista | 59.4 | 17.93 | 16 | May 1984–Dec 2011 | 1463 | 2.6 ± 1.1 |
| Harku | 59.39 | 24.58 | 36 | Jan 2003–Dec 2011 | 465 | 2.2 ± 1.0 |
| Risoe | 55.69 | 12.10 | 9 | Jun 1986–Dec 2011 | 1400 | 3.0 ± 1.4 |
| Utena | 55.5 | 25.6 | 111 | Feb 2002–Dec 2011 | 169 | 3.4 ± 1.6 |
| Clonskeagh | 53.3 | −6.23 | 43 | Feb 2007–Oct 10 | 67 | 2.7 ± 1.1 |
| Berlin | 52.53 | 13.42 | 50 | Feb 1984–Dec 2011 | 406 | 3.1 ± 1.1 |
| Braunschweig | 52.25 | 10.50 | 84 | Jan 1982–Dec 2011 | 336 | 3.1 ± 0.9 |
| Bilthoven | 52.11 | 5.18 | 4 | Feb 1987–Dec 2011 | 1048 | 3.5 ± 1.0 |
| Bruxelles | 50.84 | 4.35 | 34 | Feb 1986–Dec 2011 | 271 | 3.3 ± 1.5 |
| Offenbach | 50.1 | 8.77 | 107 | Jan 2001–Dec 2011 | 503 | 3.4 ± 1.7 |
| Praha | 50.09 | 14.42 | 202 | Jan 2002–Dec 2011 | 553 | 3.7 ± 1.2 |
| Luxembourg | 49.63 | 6.13 | 280 | Jan 1987–Dec 2011 | 1159 | 3.2 ± 1.7 |
| Vienna | 48.22 | 16.35 | 193 | May 1982–Dec 2011 | 1528 | 3.9 ± 1.6 |
| Freiburg | 48.20 | 7.87 | 411 | Jan 1989–Dec 2011 | 1115 | 4.0 ± 2.5 |
| Bratislava | 48.17 | 17.17 | 133 | Mar 2004–Dec 2011 | 59 | 5.1 ± 1.1 |
| Muenchen | 48.13 | 11.59 | 530 | Feb 1987–Jan 2009 | 264 | 3.4 ± 1.6 |
| Budapest | 47.5 | 19.11 | 117 | Jan 1987–Dec 2006 | 377 | 3.2 ± 1.5 |
| NRIRR, Budapest | 47.43 | 19.03 | 138 | Jan 2007–Dec 2011 | 113 | 4.0 ± 1.6 |
| Ljubljana | 46.09 | 14.59 | 281 | Feb 2003–Dec 2011 | 118 | 3.7 ± 1.3 |
| Milano | 45.47 | 9.18 | 125 | Feb 1988–Jan 2011 | 473 | 3.0 ± 1.1 |
| Bilbao | 43.17 | −2.94 | 380 | July 2000–Dec 2011 | 609 | 3.1 ± 1.3 |
| Seyne-sur-mer | 43.08 | 5.88 | 16 | May 1988–Jan 2007 | 212 | 4.9 ± 2.1 |
| Sofia | 42.75 | 23.33 | 522 | Aug 2003–Dec 2011 | 122 | 4.2 ± 1.2 |
| Barcelona | 41.38 | 2.12 | 52 | Jan 2001–Dec 2011 | 581 | 3.8 ± 2.0 |
| Brindisi | 40.65 | 17.95 | 11 | Feb 1988–Dec 1995 | 95 | 5.4 ± 1.5 |
| Madrid | 40.45 | −3.69 | 715 | Jan 1998–Dec 2011 | 720 | 3.8 ± 1.5 |
| Sacavem | 38.72 | −9.13 | 87 | Feb 1991–Dec 2011 | 305 | 3.7 ± 1.8 |
| Sevilla | 37.39 | −6.01 | 8 | Oct 2000–Dec 2011 | 583 | 4.0 ± 1.4 |
| Laguna | 28.46 | −16.29 | 358 | Jan 2007–Dec 2011 | 266 | 4.8 ± 1.5 |
| *Krakow* | *50.05* | *19.92* | *202* | *Dec 2002–Dec 2011* | *10* | *3.4 ± 0.8* |
| *Warszaw* | *52.23* | *21.02* | *113* | *Feb 1986–Dec 2011* | *31* | *3.3 ± 0.9* |

month (e.g. January). Using this criterion, the number of stations decreased from 34 to 32, not considering Krakow and Warsaw sites in this study (at the bottom and in italic in Table 1) due to both stations register $^{7}$Be concentrations in yearly basis.

## 4. Site description

The 32 sampling locations distributed over EU territory considered in this work are illustrated in Fig. 1. Their location (urban, rural, etc.) and topographic surroundings varies considerably ranging from stations sited in cities, on mountains or along coastal borders. In addition, sampling sites are located over a broader range of altitudes. The elevation data for each station has been estimated using the values extracted from the Digital Elevation Model over Europe called EU-DEM, from the GMES RDA project (EU-DEM, http://www.eea.europa.eu/data-and-maps/data/eu-dem), which is a 3D raster dataset with elevations captured at 1 arc second postings (2.78E-4°) that means a spatial resolution of about 30 m. From these points of view, the station locations are considerably heterogeneous, and therefore, this mosaic of monitoring stations ensures a high degree of representativeness of the results obtained in this work.

The complexity lies not only in the topographic surroundings, but also in the influence of spatial and temporal variability on similar or different atmospheric conditions. Influences from pressure systems together with the orographic characteristics of the European continent produce a high climatic variability with complicated wind patterns, different rainfall regime, gradient of temperature, etc. that affect the $^{7}$Be activity concentration.

## 5. Results

Table 1 shows a summary of the data for each station with the mean values and the corresponding standard deviation (SD). The mean value of $^{7}$Be activity concentration considering all the data available in each station ranges from 2.0 to 5.4 mBq/m$^{3}$, with an average value of 3.1 mBq/m$^{3}$. This value is comparable with that of 3 mBq/m$^{3}$ reported on $^{7}$Be activity concentration in air (UNSCEAR, 2008).

### 5.1. ANOVA statistical analysis

The first step in the data analysis was to quantitatively evaluate the fraction of the variance of $^{7}$Be activity concentration which can be ascribed to certain parameters. In other words, we wanted to define the reduction of the variance which is obtained by grouping the deterministic part associated with certain parameters. For this study all $^{7}$Be data available were used (21851 data) considering data grouped by spatial (stations) and temporal (yearly, seasonal and monthly) basis.

The ANOVA analysis of the data was performed with STATISTICA software (STATISTICA 7). In Table 2 the percentage of the variation of $^{7}$Be concentrations explained respectively by station, year, season and month are reported. The effect of each variable was analysed separately, and for all tests, $p < 0.05$ was considered to be significant.

ANOVA results showed that the percentage variation due to spatial parameter (39% explained by station) was much higher than those due to temporal ones, all of them below 10%. As expected, the

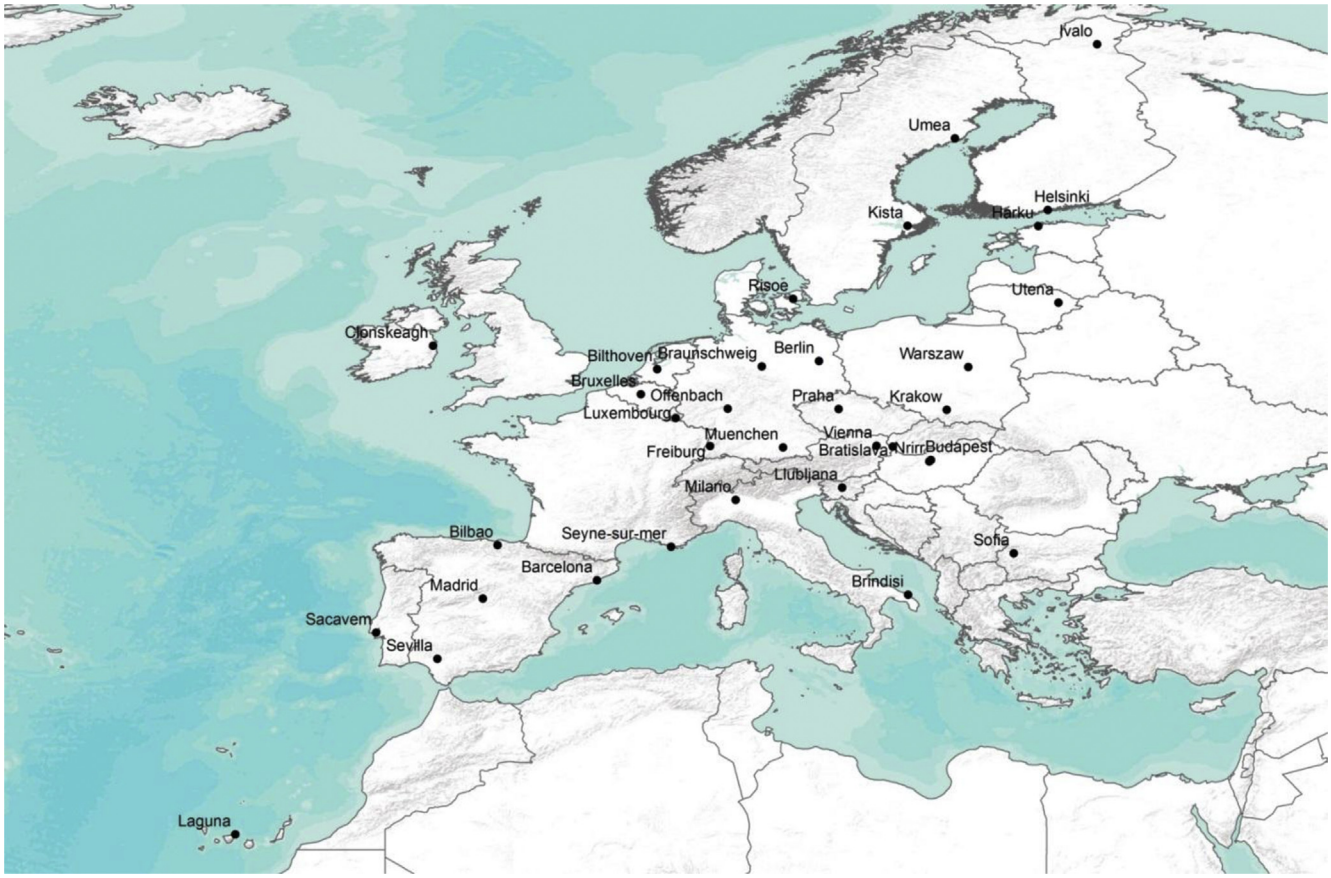


**Fig. 1.** Map of the 34 REM sample locations that provide data on $^{7}$Be concentrations in airborne particulates for the REM sparse network.

results indicate an increase of the explained $^{7}$Be activity concentrations with increasing temporal coverage. From year to month the percentage of variability increases from 3.4 % to 9.1 %. These results indicated that the temporal and spatial distributions in $^{7}$Be activity concentration in Europe were mainly caused by the characteristics of the siting, location where sampling sites are located, due to determine the impact of synoptic and regional atmospheric conditions on local scale.

In the light of the ANOVA results, the analysis of the $^{7}$Be database for the EU, presented in the following sections, has been developed considering each station separately.

### 5.2. $^{7}$Be activity concentrations in EU

Fig. 2 presents frequency distributions of $^{7}$Be activity concentrations in each sampling site over Europe. The stations are ordered by latitude from high (left side) to low (right side) (Table 1). A box-and whisker representation is often used in exploratory data analysis, to show the shape of the distribution, its central value, and its variability. This methodology is widely used to have a first overview of large amounts of data (Solazzo et al., 2012; Tukey, 1977).

**Table 2**
Percentage of the variation of $^{7}$Be activity concentrations explained by different basis.

| Source of variation | | Percentage of variation |
|---|---|---|
| Spatial basis | Stations | 39.0% |
| Temporal basis | Yearly | 3.5% |
| | Seasonal | 7.1% |
| | Monthly | 9.1% |

The results of the frequency distribution indicated the existence of a large variability in $^{7}$Be activity concentrations in Europe, underlined also by ANOVA analysis. In all sampling stations, a positive asymmetrical distribution (positively skewed) of the values was shown, as the upper quartile (P75) is farther from the median than the lower on (P25). This fact confirms the greater variability observed from P50 onwards than for lower values (P50 downward). This resulted also observing that the mean is always larger than the median, denoting the dominance of low $^{7}$Be values as well as the large impact of occasional high measures. This asymmetrical distribution is also observed in previous studies, such as Lee et al., 2007 or Tositty et al., 2014.

Regarding the value of the inter-quartile range (P75–P25), we also observed the dependence of the P75–P25 value on the latitude (the stations are listed from high to low latitude). In general, this interquartile range tended to increase with decreasing latitude. The higher is the latitude, the lower is the range. In fact, a large difference among stations located at high latitudes, such as Ivalo, Umea or Helsinki, and those at lower ones, such as Laguna or Seville, is observed.

In addition, we did not identify a clear tendency for $^{7}$Be activity concentration to increase with altitude, as previously observed (Bourcier et al., 2011). Combining the results of the box-plot with the corresponding altitudes (Table 1) we observed that the highest concentrations were not associated with the high-altitude sites. This result was caused by the fact that all the stations are located within the atmospheric boundary layer (ABL), with a depth varying largely amongst different regions of the world but is typically in the

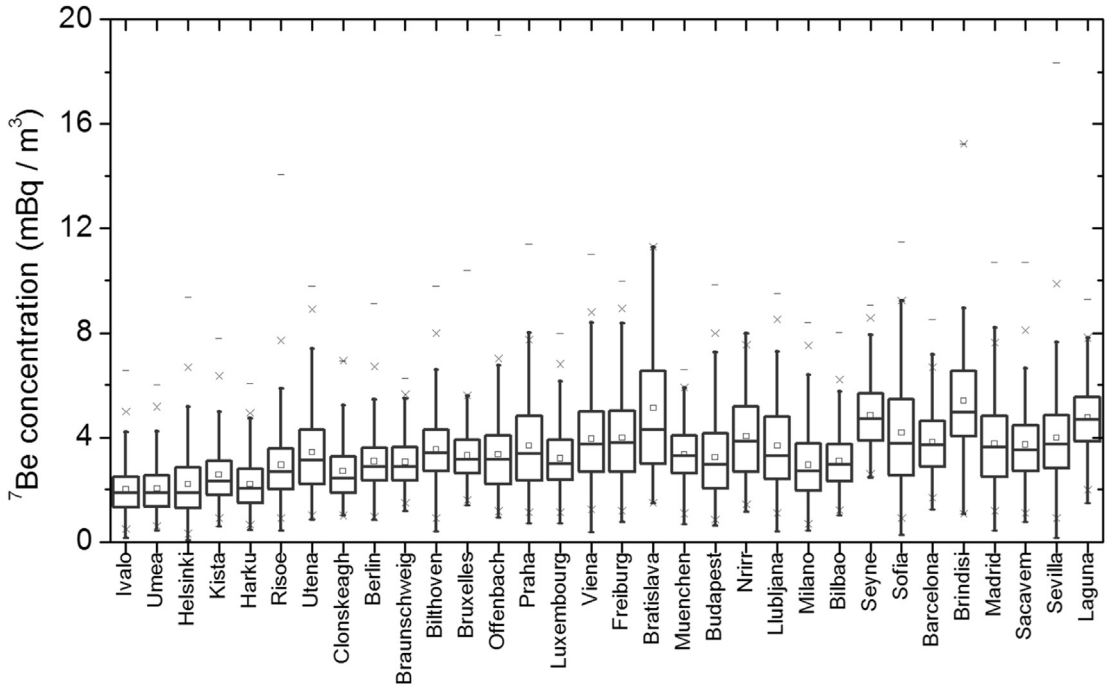


**Fig. 2.** Box-plots of $^7$Be frequency distribution at different sampling stations in the EU for the whole sampling period in each one. The rectangle represents the 50% of data (inter-quartile range from 25th to 75th percentile), the small square identifies the mean, the continuous horizontal line inside the rectangle identifies the median (50th percentile), the crosses identify the 1st and 99th percentiles respectively, and the whiskers extend between the minimum and maximum values.

order of 1–2 km over land in mid-latitudes (Stull, 1988). This layer acts as a key length scale in weather, climate, and air quality analysis to determine turbulence mixing, vertical diffusion, convective transport and cloud formation (Garratt, 1992). The intensity of these atmospheric processes present a large spatial and temporal variability, influenced also by the geographical characteristics of the sampling sties, and therefore, justify the non-existence of a well-established relationship between altitude and $^7$Be activity concentration.

### 5.3. Seasonal and monthly evolution

The seasonal variability in surface air of $^7$Be concentration in each station is displayed in Fig. 3. We point out the comparable

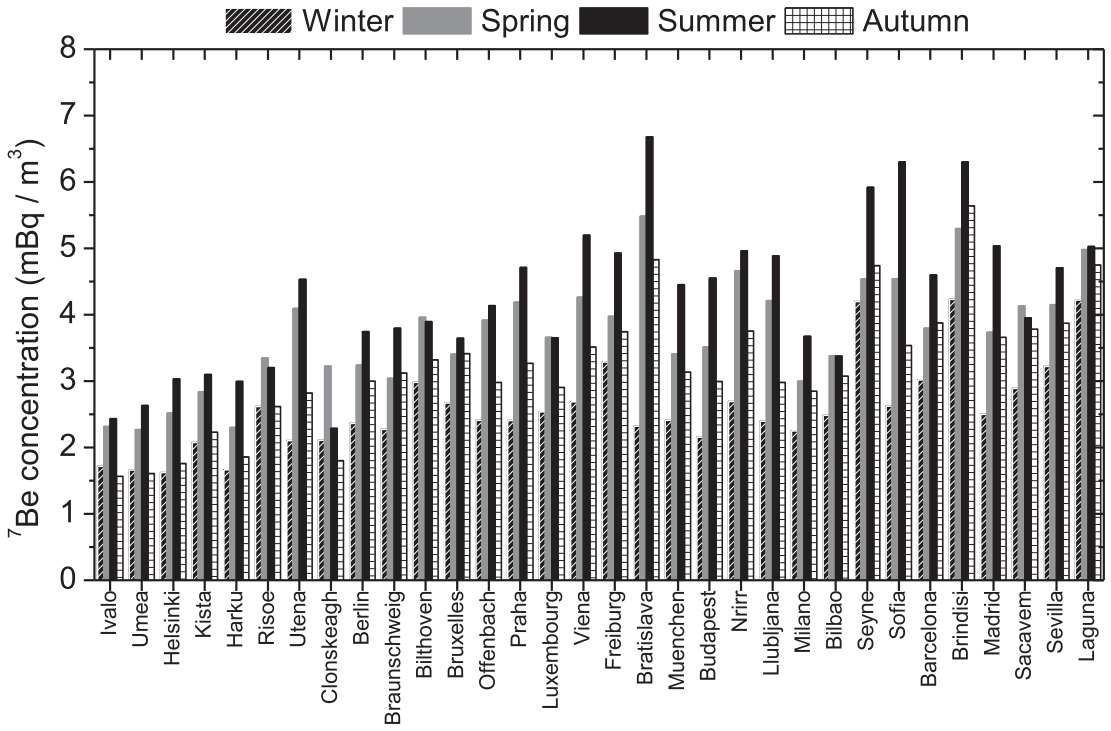


**Fig. 3.** Seasonal average of $^7$Be at the different sites.

observed trends in all stations: increasing activity in spring and summer and decreasing in winter and autumn. Therefore, this indicated the presence of a strong seasonal pattern for $^{7}$Be. The same $^{7}$Be seasonal pattern was previously observed and discussed in other studies, such as in Dueñas et al., 2001; Azahar et al., 2003; Lozano et al., 2011 in Spain, Likuku, 2006 in Ireland and Bourcier et al., 2011 in France.

This figure also marked again the latitudinal impact on $^{7}$Be activity concentrations. This influence was observed in all seasons, especially on summer. Baskaran (1995) suggested three reasons behind the seasonal variations in the surface air $^{7}$Be concentrations: 1) seasonal variations in the amount of precipitation, 2) increased stratosphere-to-troposphere exchanged during the late winter and early spring, and 3) increase vertical transport of $^{7}$Be from the upper troposphere to the middle and lower troposphere due to the decreased stability of the troposphere during the summer months. Leppänen et al., 2010 and Bourcier et al., 2011 have demonstrated the positive impact of the seasonal variation of the sun's cycle (cosmic rays) and the stratosphere-to-troposphere exchanges (there is a seasonal thinning of the tropopause and more intensity of vertical mixing in the atmosphere during the warm months). In this line, Ionnidou et al., 2014 has demonstrated the strong positive correlation between the seasonal changes of $^{7}$Be concentration in surface air and the tropopause height and temperature, confirming that the increased rate of vertical transport within the tropopause is in favour to limit the arrival of air masses enriched in $^{7}$Be to surface layers. The impact of precipitation variability has also been studied (Lee et al., 2002; Dueñas et al., 2001; Lozano et al., 2011) as the dominant removal mechanism of $^{7}$Be is washout by precipitation (McNeary and Baskaran, 2003).

To confirm the impact of these effects in the temporal distribution of air surface $^{7}$Be concentrations, Fig. 4 displays the annual distribution of the maximum and minimum monthly values in EU. This figure represents the total number of stations in which the monthly maximum or minimum values of $^{7}$Be activity concentration have been registered in the same month. We note the clear distribution of the maxima values in spring and summer (from April to September, with maximum in May and June), while the minima are registered in the cold period (from October to February, with a maximum in December). However, no clear correlation between latitude and the monthly distribution of maximum and minimum values was observed. This fact indicates an impact of the characteristics of the site and local atmospheric conditions rather than latitude in the temporal distribution of maximum and minimum monthly values.

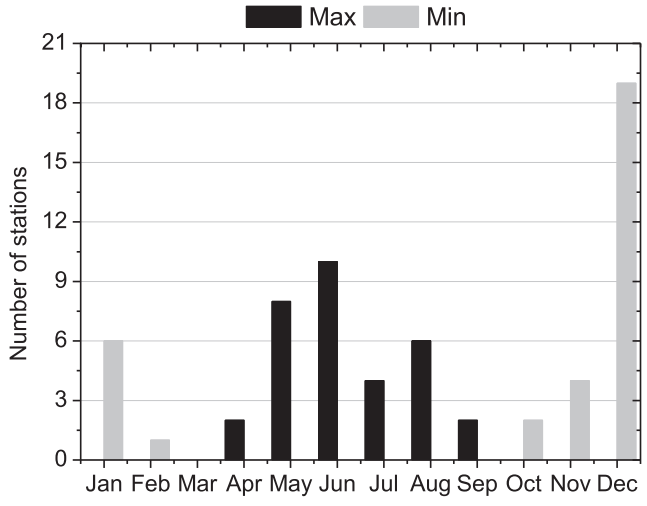


**Fig. 4.** Number of sampling stations that have been registered the monthly maximum or minimum values of $^{7}$Be activity concentration in each month.

### 5.4. Yearly average concentrations

Annual average values of the $^{7}$Be concentrations were obtained for the REM network on a yearly basis. Fig. 5 shows the yearly arithmetic means (AM) of the obtained values (different types of marks) and the total average (big black crosses) during the whole measuring period at all the stations. It is necessary to remark that stations are listed from high to low latitudes and not all of them presented the same number of years (Table 1).

No clear inter-annual tendency is found but in this figure we observe a spatial trend, showing a tendency for the mean $^{7}$Be value to increase with decreasing latitude, and therefore, confirming once more the impact of this parameter in the production of $^{7}$Be. In the same line, a reduced yearly variability with decreasing latitude is also observed. In this sense, the latitudinal effect on the $^{7}$Be yearly average concentrations causes a 3.4 mBq/m$^{3}$ variation among the considered stations, ranging from the minimum value equal to 2.0 mBq/m$^{3}$ in Ivalo and Umea to the maximum concentration of 5.4 mBq/m$^{3}$ in Brindisi (Table 1). Considering the extreme values it would be possible to calculate a latitudinal gradient of $^{7}$Be equal to 0.1 mBq/m$^{3}$ per degree.

The stations located in similar latitudes in regions of Central Europe presented low variation of $^{7}$Be mean values. The differences among them are mainly caused by the geographical location of each station, which determines the impact of the regulating processes of the variability in activity of $^{7}$Be in surface air, such as production, transport and deposition.

### 5.5. Influence of solar cycle on $^{7}$Be activity concentrations

The impact of the 11-y solar modulation on the $^{7}$Be concentrations in air is well known (e.g. Vernova et al., 2003; Talpos et al., 2005; Usokin and Kovaltsov, 2008; Leppänen et al., 2010). As $^{7}$Be is produced in the atmosphere through interaction of cosmic rays with atmospheric molecules, its production rate varies with solar modulation of galactic cosmic rays invading the heliosphere (Masarik and Beer, 1999), which is controlled by the solar magnetic field and, in turn, by solar activity.

With the aim to show the impact of the 11-y modulation on the $^{7}$Be air concentrations in Europe, Fig. 6 shows the yearly evolution from 1984 to 2011 of both $^{7}$Be activity concentrations in air at sampling stations in the EU and sunspot number (http://sidc.oma.be/silso/datafiles). This period comprises the 22nd (1986 September–1996 May), 23rd (1996 May–2008 January) and the beginning of 24th (2008 January – present) solar cycles.

This plot displays the well-known opposite, asynchronous profiles of both evolutions, following previous studies as Steinmann et al., 2013, Kikuchi et al., 2009 and Cannizzaro et al., 2004. It also showed a decreasing (increasing) trend of the solar activity ($^{7}$Be activity concentration) corresponding to the last solar cycles. The maximum average concentrations of $^{7}$Be in Europe are 3.4 mBq/m$^{3}$ in 1988, 3.7 mBq/m$^{3}$ in 1993 and 4.2 mBq/m$^{3}$ in 2009. The $^{7}$Be concentration profile showed an increase in the following three stages: 1) from 1984 to 1986, 2) from 1991 to 1993 and 3) from 2000 to 2009. We then looked for periods in which the tendency of $^{7}$Be yearly evolution is to decrease while still presenting humps and relative peaks. Later, we observed a sharp decrease for a couple of years in $^{7}$Be concentrations until it reaches the minimum activity concentrations. This evolution indicates that the yearly variation in the $^{7}$Be concentration is not a simple increase following by a decrease.

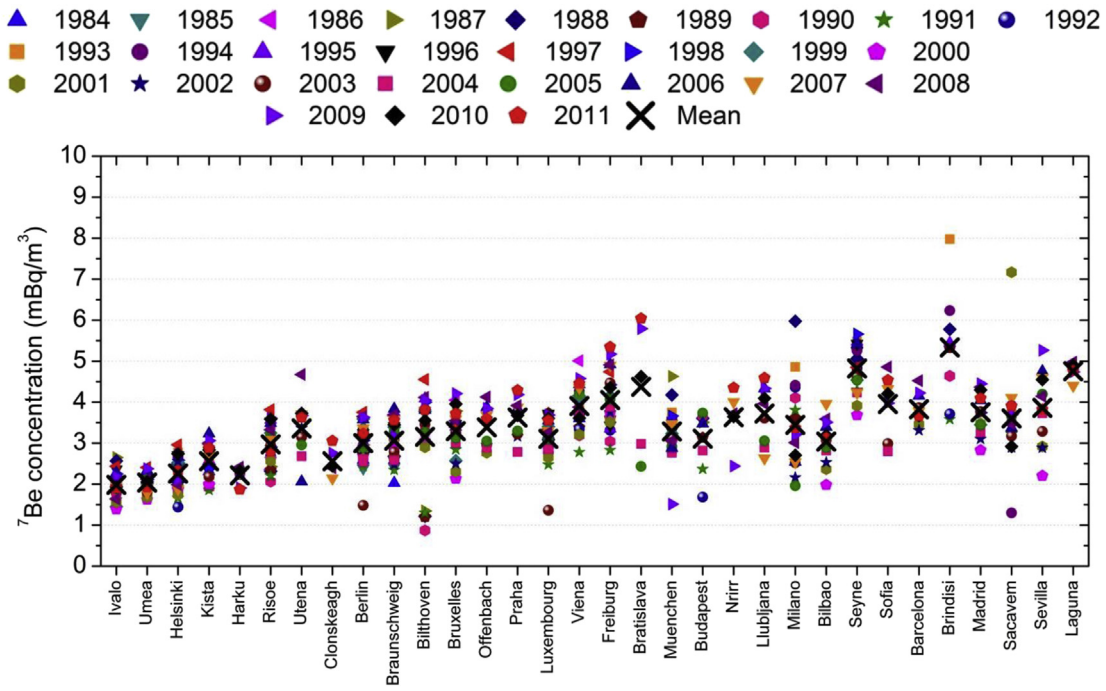


**Fig. 5.** Average of the $^{7}$Be at the different sites each year and the total average one (big black cross). Detailed yearly variability in each station can be consulted in the Annual Monitoring Report (http://rem.jrc.ec.europa.eu/RemWeb/Reports.aspx).

Once we have presented the influence of the cycles of the sun's rotation on the time variation in the whole mean $^{7}$Be concentration over Europe, we investigated the variations in the $^{7}$Be concentrations station by station (Fig. 7). Gerasopoulous et al., 2003 showed that the impact of solar cycles is not the same in each station due to different meteorological influences. To verify this fact, we have taken as reference the time period of 1999–2001 as the maximum of the 23rd solar cycle and the 2007–2009 time periods as the minimum of the 24th solar cycle. The selection of these periods was based on the availability of data in each sampling site, considering only stations not presenting any yearly gaps in each period.

Years corresponding to the maximum and minimum $^{7}$Be air concentrations during 1999–2009 were identified (not shown), indicating that the maximum values of $^{7}$Be in surface air were mainly registered during the minimum solar cycle (2007–2009) while the minimum values were observed during the maximum solar cycle (1999–2001). On the other hand, the arithmetic mean surface air concentrations during each period were calculated (Table 2). These results showed that the Helsinki and Muenchen data were the only ones showing decrease in the mean surface air concentrations, this difference decreases with increasing of latitude. The other stations presented an increase ranged from the 0.2 mBq/m$^3$ at Seyne-sur-mer to 1.4 mBq/m$^3$ at Bruxelles (Fig. 7). In general, these variations agree with the results reported in Kulan et al., 2006 which established the effect of solar modulation to surface air $^{7}$Be concentrations in mid-latitude in a range of 30–40 % while at higher latitudes (55–68 N) the effect was ranged between 15 and 20 %. Considering these results we can suggest that yearly surface air $^{7}$Be concentrations in Europe are highly dependent on solar modulation rather than atmospheric effects. The latter has stronger impact for the monthly data, in terms of the impact of local meteorological conditions affecting the amount of precipitation,

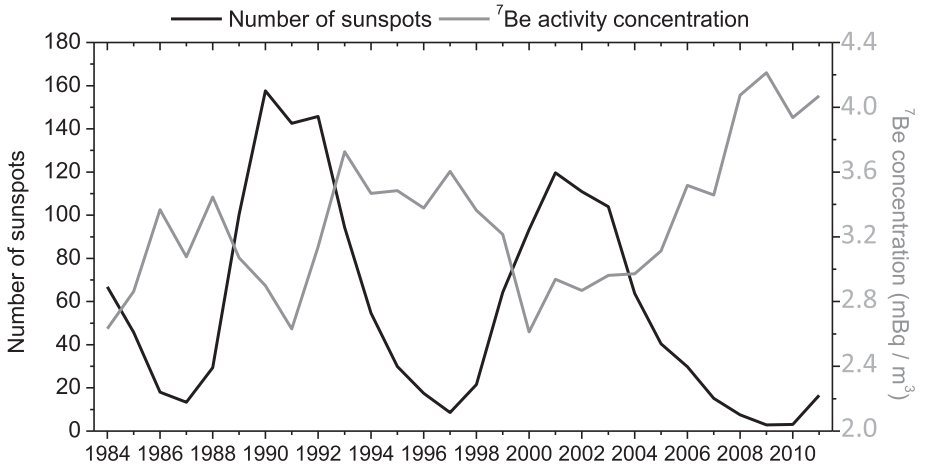


**Fig. 6.** Yearly variations in the $^{7}$Be activity concentration and sunspot number from 1984 to 2011.

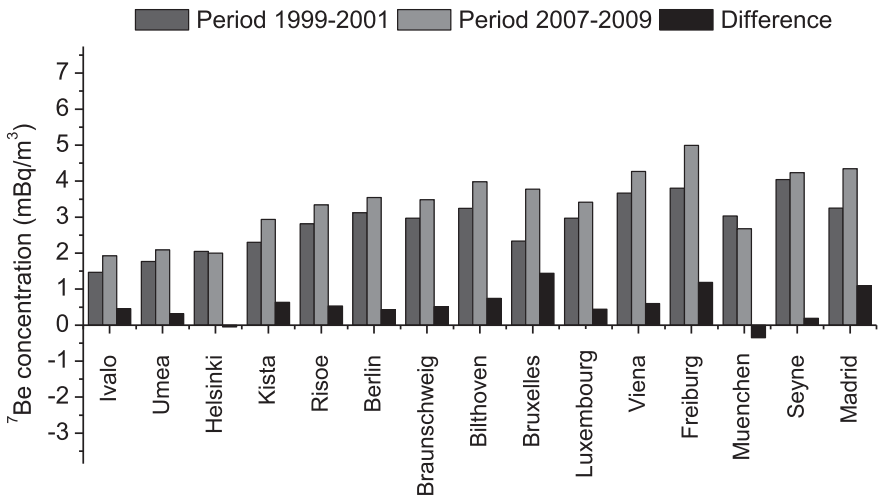


**Fig. 7.** The average of $^{7}$Be activity for each maximum (1999–2001) and minimum (2007–2009) solar cycle and the difference between both periods calculated using arithmetic mean.

and variations in large-scale atmospheric circulation or vertical transport of the air.

## 6. Conclusions

In this paper, an analysis of airborne $^{7}$Be activity concentrations in air measured at 34 different sampling stations located across the European Union, covering a range of latitudes (35°–72°N) and altitudes (20°W–40°E), was performed, using data from the 1984–2011 period. Data collected by the Radioactivity Environmental Monitoring (REM) network were used.

The ANOVA analysis showed that the percentage variation of $^{7}$Be activity concentrations connected to spatial parameter (location of the sampling sites) is much higher than that due to temporal ones (yearly, seasonal and monthly). However, while a large impact of latitude on $^{7}$Be activity concentrations was observed, no effect of the altitude on the $^{7}$Be concentrations was observed, probably due to the fact that all the stations lie in the ABL.

The results display the large impact of a 11-y modulation on the $^{7}$Be air concentrations in Europe. Different surface airborne $^{7}$Be average concentrations were observed in the time periods of 1999–2001 (maximum of the 23rd solar cycle) and 2007–2009 (minimum of the 24th solar cycle), with a large spatial variability in the observed difference.

A seasonal and monthly evolution of $^{7}$Be activity concentrations is also well observed in all stations. The maximum concentrations were observed in spring-summer (April to September), whereas the minima were registered in autumn-winter (October to February).

These results have shown the spatial and temporal variability of this natural radionuclide in the atmosphere. Due to its use as a tracer of atmospheric processes that affects concentrations of radionuclides in the Earth's surface, these results can be used as reference for modelling studies of atmospheric processes, which are important phenomena to be taken into account in the case of a nuclear accident.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2014.12.003.

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